

Translation

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PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

10/523264

(PCT Article 36 and Rule 70)

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| Applicant's or agent's file reference B02/0402PC | FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416) | |
| International application No. PCT/EP2003/008478 | International filing date (day/month/year) 31 July 2003 (31.07.2003) | Priority date (day/month/year) 01 August 2002 (01.08.2002) |
| International Patent Classification (IPC) or national classification and IPC B01J 31/22, 37/04, C07D 305/12, C07F 15/06 | | |
| Applicant BASF AKTIENGESELLSCHAFT | | |

- This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 6 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 3 sheets.

- This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

| | |
|---|--|
| Date of submission of the demand 27 February 2004 (27.02.2004) | Date of completion of this report 22 July 2004 (22.07.2004) |
| Name and mailing address of the IPEA/EP | Authorized officer |
| Facsimile No. | Telephone No. |

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International Application No.

PCT/EP2003/008478

I. Basis of the report

1. With regard to the elements of the international application:*

- ☐ the international application as originally filed
- ☒ the description:
pages 1-12, 14, as originally filed
pages _____, filed with the demand
pages 13, filed with the letter of 27 May 2004 (27.05.2004)
- ☒ the claims:
pages _____, as originally filed
pages _____, as amended (together with any statement under Article 19
pages _____, filed with the demand
pages 1-9, filed with the letter of 27 May 2004 (27.05.2004)
- ☐ the drawings:
pages _____, as originally filed
pages _____, filed with the demand
pages _____, filed with the letter of _____
- ☐ the sequence listing part of the description:
pages _____, as originally filed
pages _____, filed with the demand
pages _____, filed with the letter of _____

2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language _____ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of the translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages _____
- ☐ the claims, Nos. _____
- ☐ the drawings, sheets/fig _____

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**

* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rule 70.16 and 70.17).

** Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**1. Statement**

| | | | |
|-------------------------------|--------|--------|-----|
| Novelty (N) | Claims | 1-6, 9 | YES |
| | Claims | 7, 8 | NO |
| Inventive step (IS) | Claims | | YES |
| | Claims | 1-9 | NO |
| Industrial applicability (IA) | Claims | 1-9 | YES |
| | Claims | | NO |

2. Citations and explanations

- 1) The present application relates to methods of catalytic carbonylation which show a stereoselective tendency, more particularly for the production of lactones from oxiranes, and to the suitable catalysts. The catalysts concern a catalyst system comprising a carbonylation catalyst A which consists of neutral or anionic transition metal complexes that contain a central atom having 0 or negative formal charge, more particularly a Co-containing catalyst, and a chiral Lewis acid B, more particularly a (salen)Cr(III) complex.

In this context, [(salph)Al(THF)₂][Co(CO)₄] is explicitly excluded as a catalyst.

The amendments find their support in the original application, in particular the description.

- 2) The following documents are mentioned:

D1: J. AM. CHEM. SOC., Vol. 124, No. 7, (2002-02-20), pages 1174-1175

D2: WO-A-00/09463

D3: ACC. CHEM. RES., Vol. 33, No. 6, (2000-06), pages 421-431

D4: INORGANIC CHEMISTRY (1993), 32(23), 5385-90

D5: C.A. 117:263541 (BREWER)

D6: WO-A-03/050154 (CORNELL RES FOUNDATION INC)

19 June 2003 (2003-06-19).

Document D6 was published after the priority date of the present application. On the assumption that the priority is valid, that document is therefore not considered to be prior art.

3) Novelty

D1 already describes the production of lactones by catalytic carbonylation of oxiranes using $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$ as a catalyst; however, that catalyst system is excluded in the present application and a stereoselective tendency is not achieved.

Documents D2 and D3 describe the stereoselective ring opening of oxiranes using $(\text{salen})\text{Cr}(\text{III})$ complexes, but no carbonylation.

Document D4 describes Cr/Co complexes in which the corresponding central atom has a formal charge of +2.

Document D5, however, describes Cr/Co complexes, the structure of which corresponds to the structure of the present catalyst systems; the corresponding central atom would, in the examiner's opinion, have a formal charge equal to 0. That document is therefore regarded as prejudicial to the novelty of claims 7 and 8. It is noted in this connection that it is not evident that the complexes as per D5 would not show similar catalytic activity.

It should be noted that document D6 describes similar catalyst systems for conversions similar to those of the present application.

4) Inventive step/workability

Document D1 is considered to be the closest prior art. Having regard to D1, the problem appears to be that of providing stereoselective catalyst systems.

At present, the subject matter of the application appears to be an obvious solution to this problem.

A person skilled in the art would realize from the mechanism indicated in D1 that alternative combinations of Lewis acid with carbonylation catalysts can be used instead of $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$. Attention is drawn in this connection to the basis for the selection of $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$ in D1. Moreover, the $(\text{salen})\text{Cr}(\text{III})$ complexes as per D2 and D3 would be suitable candidates, because they permit similar activation of the oxirane, while offering the advantage of stereospecificity.

Should the applicant be able to argue convincingly that exchange of the Lewis acid in combination with carbonylation catalysts in order to achieve a stereoselective tendency would nevertheless not be familiar to a person skilled in the art on the basis of D1 and D2 or D3 and represented an inventive feat, the following observations pertain.

Given the scope and chosen definition of the current claims, the same person skilled in the art then still finds himself confronted by the problem of selecting suitable stereoselective Lewis acids in combination with carbonylation catalysts. The only sign of a solution proposed by the claims is the information that $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$ is not to be used. However, it was already known in the prior art that $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$ does not show a stereoselective tendency (D1). The claim therefore

effectively defines the subject matter only in terms of a problem and it is not evident therefrom how the relevant person skilled in the art could put the intended solution into practice throughout the entire scope of the claims, without thereby being inventive.

as enclosed to IPER

We claim:

1. A process for preparing lactones by catalytic carbonylation of oxiranes using a catalyst system comprising

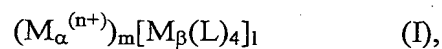
a) at least one carbonylation catalyst A comprising uncharged or anionic transition metal complexes of metals of groups 5 to 11 of the Periodic Table of the Elements, in which the central atom bears the formal charge 0 or a negative formal charge, and

b) at least one chiral Lewis acid B,

with the exception of [(salph)Al(THF)₂][Co(CO)₄], as catalyst, wherein the lactones are mixtures of S- and R-lactones having an excess of one enantiomer.

2. A process as claimed in claim 1, wherein the ligands in the carbonylation catalyst A are uncharged ligands.

3. A process as claimed in claim 1 or 2, wherein the carbonylation catalyst A used comprises transition metal complexes of the formula (I)



where

M_{β} is a transition metal of groups 8 to 10 of the Periodic Table of the Elements bearing the formal charge -1,

L is PR_3 , $P(OR)_3$, NR_3 , SR_2 , OR_2 , CO, R-CN, R-NO₂, (RO)(R'O)C=O, (R)(R')C=O, (R)C=O(OR'),

M_{α} is a metal of group 1 or 2 of the Periodic Table of the Elements, Zn or Hg, bis(triarylphosphine)iminium, trityl or $T(R)_4$ where

T is N, P or As,

R, R' are each, independently of one another, H, alkyl, aryl, alkaryl or aralkyl,

n, m are each 1 or 2 and

1 is $n \times m$.

4. A process as claimed in any of claims 1 to 3, wherein the transition metals present in the carbonylation catalyst A are Re, Co, Ru, Rh, Fe, Ni, Mn, Mo, W or mixtures thereof.

5. A process as claimed in claim 4, wherein Co is present as transition metal in the carbonylation catalyst A.

6. A process as claimed in any of claims 1 to 5, wherein the chiral Lewis acid B comprises compounds of metals of groups 2 to 13 of the Periodic Table of the Elements which are present in coordinatively unsaturated form under the reaction conditions.

7. A catalyst as defined in any of claims 1 and 2 to 6, with the exception of $[(\text{salph})\text{Al}(\text{THF})_2][\text{Co}(\text{CO})_4]$.

8. A process for preparing catalysts as claimed in claim 7 by mixing the components A and B.

9. The use of a catalyst as claimed in claim 7 in carbonylation reactions.

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